

WHAT IS CLAIMED IS:

1. A method of forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and capable of ensuring smoothness of a surface coated therewith on a substrate comprising the steps of:

(a) preparing a liquid composition containing a silicon compound obtained by hydrolyzing tetraalkyl ortho silicate (TAOS) and alkoxysilane (AS) expressed by the following general formula (I) in the presence of tetraalkyl ammonium hydroxide (TAAOH):



wherein X indicates a hydrogen atom, a fluorine atom, or an alkyl group, a fluorine-substituted alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; R indicates a hydrogen atom, or an alkyl group, an aryl group or a vinyl group each having 1 to 8 carbon atoms; and n is an integral number from 0 to 3.

(b) applying the liquid composition on a substrate;

(c) heating the substrate at a temperature in a range from 80 to 350°C; and

(d) curing the substrate at a temperature in a range from 350 to 450 °C.

2. A method of forming an amorphous silica-based coating film with a low dielectric constant having a high film strength and excellent hydrophobic property and capable of ensuring smoothness of a surface coated therewith on a substrate comprising the steps of:

(a) preparing a liquid composition containing a silicon compound obtained by hydrolyzing or partially hydrolyzing tetraalkyl ortho silicate (TAOS) in the presence of tetraalkyl ammonium hydroxide (TAAOH), mixing the reaction product with the

alkoxysilane (AS) expressed by the general formula (I) above or a hydrolysate or a partial hydrolysate thereof, and further hydrolyzing all or a portion of the mixture according to the necessity;

(b) applying the liquid composition on a substrate;

5 (c) heating the substrate at a temperature in a range from 80 to 350°C; and

(d) curing the substrate at a temperature in a range from 350 to 450 °C.

3. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 2, wherein said tetraalkyl ortho
10 silicate (TAOS) used in the preparing step (a) is tetraethyl ortho silicate (TEOS), tetramethyl ortho silicate (TMOS) or a mixture thereof.

4. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 3, wherein said alkoxysilane (AS)
15 used in the preparing step (a) is methytrimethoxy silane (MTMS), methyltriethoxy silane (MTES) or a mixture thereof.

5. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 4, wherein said tetraalkyl ammonium
20 hydroxide (TAAOH) used in the preparing step (a) is tetrapropyl ammonium hydroxide (TPAOH), tetrabutyl ammonium hydroxide (TBAOH) or a mixture thereof.

6. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 5, wherein a content of impurities
25 comprising compounds of alkali metal elements such as sodium (Na) and potassium (K)

contained in said tetraalkyl ammonium hydroxide (TAAOH) used in the preparing step (a) is 50 ppb by weight or below on respective element bases.

7. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 6, wherein a content of impurities comprising compounds of halogen group elements such as bromine (Br) and chlorine (Cl) contained in said tetraalkyl ammonium hydroxide (TAAOH) used in the preparing step (a) is 1 ppm by weight or less on respective element bases.

8. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 7, wherein a molar ratio (TAOS/AS) of said tetraalkyl ortho silicate (TAOS) and said alkoxysilane (AS) used in the preparing step (a) is in a range from 6/4 to 2/8 in terms of SO_2 .

9. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 8, wherein a molar ratio (TAAOH)/(TAOS+AS) of said tetraalkyl ammonium hydroxide (TAAOH) and the components for forming a silica-based coating film (TAOS+AS) used in the preparing step (a) is in a range from 1/10 to 7/10 in terms of SO_2 .

10. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 9, wherein the operation used in the applying step (b) is executed with a spin coat method.

11. The method of forming an amorphous silica-based coating film with a low

dielectric constant according to any of claims 1 to 10, wherein the operation used in the heating step (c) is executed for 1 to 10 minutes in the atmosphere of nitrogen or air.

12. The method of forming an amorphous silica-based coating film with a low dielectric constant according to any of claims 1 to 11, wherein the operation used in the curing step (d) is executed for 5 to 90 minutes in the atmosphere of nitrogen.

13. An amorphous silica-based coating film with a low dielectric constant, wherein the obtained coating film using the method according to claims 1 to 12 has a specific dielectric constant of 2.5 or below and a film strength of Young's modulus of 6.0 GPa or more.

14. The amorphous silica-based coating film with a low dielectric constant according to claim 13, wherein said coating film contains pores having an average diameter of 3 nm or below and also with volume percentage of micropores each with the diameter of 2 nm or below of 70% or more.

15. The amorphous silica-based coating film with a low dielectric constant according to any of claims 13 to 14, wherein said coating film has a smooth surface with surface roughness (Rms) of 1 nm or below.

16. The amorphous silica-based coating film with a low dielectric constant according to any of claims 13 to 15, wherein said coating film is an amorphous silica-based coating film not having any X-ray diffraction peak specific to a MFI crystal structure.

17. The amorphous silica-based coating film with a low dielectric constant according to any of claims 13 to 16, wherein said coating film is an inter-layer insulation film or an inter-metal insulation film formed on a semiconductor substrate..